Nonequilibrium Zeeman-splitting in quantum transport through nanoscale junctions

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(Dated: January 10, 2013)

We calculate the differential conductance $G(V)$ through a quantum dot in an applied magnetic field. We use a Keldysh conserving approximation for weakly correlated and the scattering-states numerical renormalization group for the intermediate and strongly correlated regime out of equilibrium. In the weakly correlated regime, the Zeeman splitting observable in $G(V)$ strongly depends on the asymmetry of the device. In contrast, in the strongly correlated regime the position Δ_K of the Zeeman-split zero-bias anomaly is almost independent of such asymmetries and of the order of the Zeeman energy Δ_0. We find a crossover from the purely spin-fluctuation driven Kondo regime at small magnetic fields with $\Delta_K < \Delta_0$ to a regime at large fields where the contribution of charge fluctuations induces larger splittings with $\Delta_K > \Delta_0$ as it was observed in recent experiments.

Quantum transport through interacting nanoscale junctions has attracted much interest over the last decade[1]. In the Coulomb blockade regime single electron transfer through a quantum dot coupled to two leads is blocked due to the charging energy $U = e^2/(2C)$ of the device, where C is the capacitance[2]. For quantum dots with an odd number of electrons, spin-flip scattering opens up a new transport channel due to the Kondo effect[3]. The zero-bias conductance increases for decreasing temperature and even reaches the unitary limit value $\frac{2e^2}{h}$ characteristic of a fully transparent device[4].

Recent experiments have measured the splitting of this zero-bias anomaly (ZBA) of the differential conductance $G(V)$ in a finite magnetic field[5, 6]. While in some experiments [5] the splitting was smaller than the Zeeman splitting in accordance with theory, in others[6] a crossover to larger splitting exceeding the theoretical predictions was observed at large magnetic field. However, much of the theoretical calculations[7–9] have focused on the equilibrium situation, i.e. only the linear response conductance. Studies out of equilibrium are usually either perturbative in some quantities[11] and thus restricted to special parameter values (e.g. large or small voltages, small Coulomb interaction, etc) or employ methods which do not properly account for the low temperature Fermi liquid in equilibrium[10]. Additionally, results obtained for the Kondo-model where charge fluctuations of the quantum dot are explicitly excluded might not be directly applicable to experiments, as such fluctuations are inevitably present in actual devices.

In this paper we address the question how a combination of charge fluctuations and nonequilibrium effects in strongly correlated systems can indeed explain the experimentally observed discrepancies. We will pinpoint the crossover from spin-fluctuation to charge-fluctuation driven transport with increasing bias and fields. Additionally, we show that the bias dependence of the nonequilibrium spectral function yields a splitting of the ZBA in the Kondo regime which is robust against asymmetries in contrast to a single-particle picture used in mean-field descriptions.

We employ a Keldysh based conserving approximation in the weakly correlated regime[12, 13] and the scattering-states numerical renormalization group (SNRG)[13] in the strongly correlated regime. Both methods are applicable to arbitrary bias V and reduce to the corresponding equilibrium theory for $V \to 0$. The SNRG being non-perturbative covers the full range of interactions but is numerically much more expensive. Its accuracy is determined by discretization errors[14]. Since the SNRG coincides with the Keldysh approach at small U the error remains well controlled as demonstrated in Ref. [13]. The temperature is set much smaller than any characteristic energy scale of the problem and is thus considered as $T \to 0$.

Since the Kondo temperature T_K is exponentially dependent on U[3], devices must be operated[1, 5, 6] in an intermediate regime with a moderate U to observe the Kondo effect experimentally. Consequently, spin and charge fluctuations are not well separated. The latter become significant at large bias and lead to a modification of the splitting as shown below. We include the charge fluctuations by considering the single impurity Anderson model (SIAM) comprising a single orbital on the quantum dot coupled to two leads,

$$\mathcal{H} = \sum_{\sigma,\alpha=L,R} \int d\epsilon \, (\epsilon - \mu_\alpha) \, c^\dagger_{\epsilon,\sigma\alpha} c_{\epsilon,\sigma\alpha} + \sum_{\sigma=\pm 1} E_\sigma \hat{n}_\sigma \quad (1)$$

$$+ U \hat{n}_\uparrow \hat{n}_\downarrow + \sum_{\alpha\sigma} \sqrt{\frac{\Gamma_\alpha}{\pi}} \int_{-D}^{D} d\epsilon \left\{ d^\dagger_\sigma c_{\epsilon\sigma\alpha} + c^\dagger_{\epsilon\sigma\alpha} d_\sigma \right\}.$$

d_σ and $d^\dagger_\sigma$ ($c_{\epsilon\sigma\alpha}$ and $c^\dagger_{\epsilon\sigma\alpha}$) are the annihilation and creation operators for electrons on the quantum dot (in lead $\alpha = L, R$) with spin $\sigma = \pm 1$, $\hat{n}_\sigma = d^\dagger_\sigma d_\sigma$, and U is the charging energy of the dot. We allow for different coupling Γ_α of lead α to the dot, but fix the total coupling strength, $\Gamma = \Gamma_L + \Gamma_R$, which is used as the unit of energy. The asymmetry of the coupling is characterized by

the ratio $R = \Gamma_R/\Gamma_L$. In a finite magnetic field H the single-particle levels of the quantum dot are shifted by the Zeeman energy $\Delta_0 = g\mu_B H$, $E_\sigma = E_d + \frac{\sigma}{2}\Delta_0$. For simplicity, we use a constant density of states between $-D$ and D for the noninteracting lead electrons in the wide-band limit $D \gg U, |E_\sigma|, \Gamma_\alpha$.

The total voltage drop across the junction is given by the difference in the chemical potentials of the two leads, $V = \mu_L - \mu_R$. In order to model the individual voltage drops at each contact we employ a serial resistor model, where the voltage at each contact is inversely proportional to its coupling strength. Keeping the dot-levels voltage independent, the chemical potentials in the leads are then given by $\mu_L = \frac{R}{1+R}V$ and $\mu_R = -\frac{1}{1+R}V$. Two important limits are incorporated. (i) For symmetric coupling ($R = 1$) half the voltage drops at each side, and nonequilibrium effects are most pronounced. (ii) In the tunneling regime (R or $1/R \to 0$) only an infinitesimal current flows through the junction. The quantum dot is in equilibrium with the stronger coupled lead and resembles an usual SIAM[3], where the weaker coupled lead acts only as a probe similar to a STM tip.

The current I through the quantum dot is determined by the nonequilibrium spectral function $\rho_\sigma(\omega, V)$ [15]

$$I = \frac{G_0}{e} \sum_\sigma \int d\omega \, [f_L(\omega) - f_R(\omega)] \, \pi\Gamma\rho_\sigma(\omega, V), \quad (2)$$

where $G_0 = \frac{e^2}{h}\frac{4R}{(1+R)^2}$, and $f_\alpha(\omega) = \frac{1}{1+e^{\beta(\omega-\mu_\alpha)}}$ are the Fermi functions. The differential conductance $G(V) = dI/dV$ reveals the Zeeman-splitting of the quantum dot levels in a finite magnetic field. The peak positions in $G(V)$ depend on the energetic level position of the dot, denoted by $\epsilon_{\rm dot}$, as well as on the coupling asymmetry R.

In the noninteracting case the dot spectral function remains voltage independent. Thus, the differential conductance at zero temperature and arbitrary R is given via the *equilibrium* spectral function[16]

$$G_{\rm Eq}(V) = \frac{\pi\Gamma G_0}{1+R} \left[\rho_{\rm Eq}(\mu_R) + R\,\rho_{\rm Eq}(\mu_L) \right], \quad (3)$$

and maxima in $G_{\rm Eq}(V)$ emerge when one chemical potential coincides with a dot level, $\mu_\alpha \approx \epsilon_{\rm dot}$. For $R > 1$, the dominant contribution comes from the second term in Eq. (3) and a pronounced peak occurs for voltages $V_{\rm max} = (1 + \frac{1}{R})\epsilon_{\rm dot}$. For symmetric coupling $R = 1$, where both terms contribute equally, a factor of two between peak positions in $G_{\rm Eq}(V)$ and $\rho_{\rm Eq}(\omega)$ results, $V_{\rm max} = 2\epsilon_{\rm dot}$, while in the tunneling regime $R \to \infty$, $G_{\rm Eq}(V) \propto \rho_{\rm Eq}(\omega = V)$.

This behavior extends to the weakly correlated regime. The differential conductance in a finite magnetic field $\Delta_0 = 2\Gamma$ is shown in Fig. 1 for various values of R and a moderate Coulomb interaction $U = -2E_d = \Gamma$. For comparison, we added the corresponding equilibrium estimation $G_{\rm Eq}(V)$ as thin black dashed lines. Even though
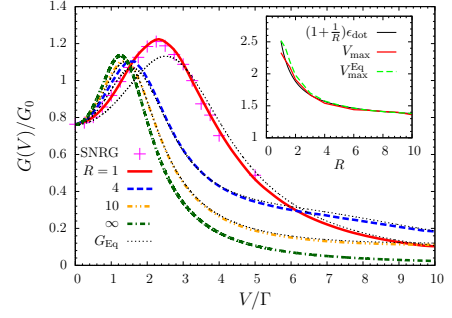

FIG. 1. (Color online) Normalized differential conductance $G(V)/G_0$ of a particle-hole symmetric quantum dot ($E_d = -U/2$) with moderate Coulomb interaction $U = \Gamma$ in a finite magnetic field $\Delta_0 = 2\Gamma$ and various coupling asymmetry parameters R as function of bias voltage V. The curves are calculated in self-consistent second Born approximation within the Keldysh formalism[13]. For comparison, the crosses are SNRG result for $R = 1$. The estimates $G_{\rm Eq}(V)$ are also shown as thin dashed black lines for each R. The inset shows the position of maxima as function of R.

there are pronounced deviations for $R = 1$ due to the U-driven voltage dependence in $\rho_\sigma(\omega, V)$, there is an overall agreement between the equilibrium estimate $G_{\rm Eq}(V)$ and the true nonequilibrium conductance $G(V)$. Both results quickly converge upon increasing R and agree almost perfectly already for $R = 10$.

Additional to the bare Zeeman splitting $\epsilon_{\rm dot}^{U=0} = \pm\frac{\Delta_0}{2}$ for a noninteracting dot, the self-energy corrections[13] caused by the finite U shift $\epsilon_{\rm dot}^{U>0}$ to slightly larger values. For the parameter values of Fig. 1 the peak positions in $\rho_{\rm Eq}(\omega)$ are $\epsilon_{\rm dot} \approx \pm 1.25\Gamma$. As visible in the inset, the maxima in $G(V)$ as well as $G_{\rm Eq}(V)$ are located approximately at $V_{\rm max} \approx (1 + \frac{1}{R})\epsilon_{\rm dot}$, and notable deviations occur only near symmetric coupling. The second contribution from the weaker coupled lead occurs at $V_{\rm max} \approx (R + 1)\epsilon_{\rm dot}$ and is visible for $R = 4$ as the small shoulder at $V \approx 6.5\Gamma$.

Fig. 2(a,b) shows the nonequilibrium conductance of a strongly correlated quantum dot in a finite magnetic field with particle-hole symmetric and asymmetric single-particle energy for various R. Only positive voltages are shown for the particle-hole symmetric dot of panel (a) due to the symmetry $G(-V) = G(V)$. The Zeeman energy considerably exceeds the corresponding Kondo temperatures, $T_K \approx 0.09\Gamma$ for $E_d = -4\Gamma$ and $T_K \approx 0.2\Gamma$ for $E_d = -2\Gamma$. In both cases, the tunneling regime is perfectly approached upon increasing R, $G(V, R \to \infty) \propto \rho_{\rm Eq}^{\rm NRG}(V)$ which is added to the plots as black lines.

We clearly can distinguish two voltage regimes. At large voltages $G(V)$ strongly depends on E_d as well as on the asymmetry R. The large-voltage maximum is reminiscent of the atomic charge excitation of the dot. Its position $V_{\rm max}$, as indicated by the arrows in Fig. 2(a,b) and plotted in Fig. 2(c), is again consistent
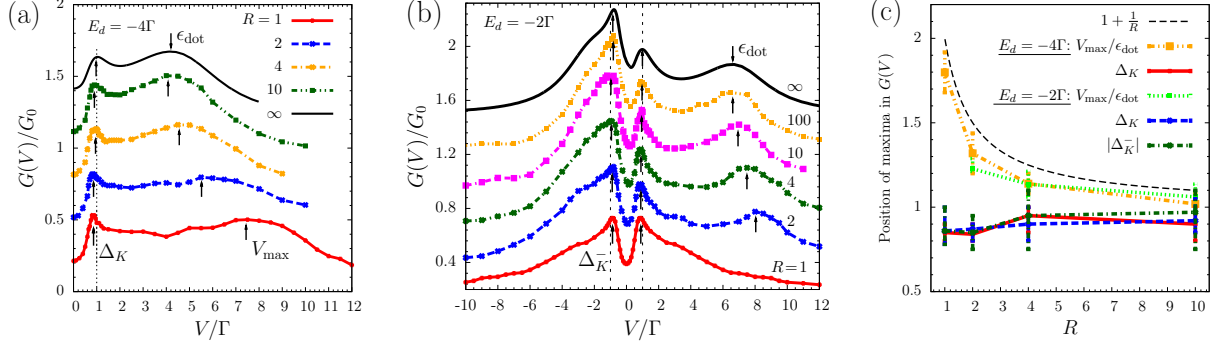
FIG. 2. (Color online) Panels (a) and (b): Normalized differential conductance calculated with SNRG for large Coulomb interaction $U = 8\Gamma$ in a finite magnetic field, $\Delta_0 = \Gamma$, and for various coupling asymmetries. (a) $E_d = -4\Gamma$ (b) $E_d = -2\Gamma$. The vertical dashed lines indicate the Zeeman-energy Δ_0 and the different curves are offset by a constant. The small arrows indicate the position of maxima in $G(V)$, which are plotted in panel (c) as function of R. Δ_K^- denotes the position at negative voltages of panel (b).

with $V_{\max} \approx (1 + \frac{1}{R})\epsilon_{\mathrm{dot}}$, where $\epsilon_{\mathrm{dot}} \approx 4.2\Gamma$ ($\epsilon_{\mathrm{dot}} \approx 6.6\Gamma$) for $E_d = -4\Gamma$ ($E_d = -2\Gamma$) has been obtain from the equilibrium NRG spectral function $\rho_{\mathrm{Eq}}^{\mathrm{NRG}}(\omega)$.

In contrast, the position of the Zeeman-split ZBA Δ_K at low voltage (also indicated by arrows) appears to be almost *independent* of particle-hole and coupling asymmetry as clearly seen in Fig. 2(a,b). Δ_K only slightly increases with increasing R [see Fig. 2(c)] and is essentially given by the Zeeman-energy $\Delta_K \approx \Delta_0$. In the linear response regime[7–9] the spectral function is approximated to be voltage independent: $\rho(\omega, V) \approx \rho_{\mathrm{Eq}}(\omega)$. This would imply that the maximum of the Zeeman-split ZBA in $G(V)$ should occur at $\Delta_K \approx (1 + \frac{1}{R})\omega_{\mathrm{Kondo}}$, where ω_{Kondo} is the position of the Zeeman-split Kondo resonance in $\rho_{\mathrm{Eq}}(\omega)$. As a hallmark of the Kondo effect, the splitting of the Kondo resonance in $\rho_{\mathrm{Eq}}(\omega)$ is strongly enhanced and approaches *twice* the value of a noninteracting level for large fields, $\omega_{\mathrm{Kondo}} \approx \Delta_0$[7–9]. The position of the Zeeman-split ZBA would thus evolve with R from $\Delta_K \approx \Delta_0$ in the tunneling regime to $\Delta_K \approx 2\Delta_0$ for a symmetric junction. Therefore, this simple picture which partially extents to the high-voltage behavior completely fails for the Kondo-correlated ZBA!

The ZBA arises from magnetic spin-flip scattering[17], where the spin of the electron on and the spin of the electron scattered across the dot contributing to the current are both flipped. In a finite external magnetic field such a spin-flip is associated with a finite energy cost Δ_0, which must be provided by the bias voltage for the transport channel to open, $V \gtrsim \Delta_0$. Hence, the asymmetry governs only the magnitude of the peak (and might lead to a merge with a charge excitation peak as in Fig. 2(b) for negative voltages) but it does not alter its position as this is determined in leading order only by the difference in energy between the initial and final state of the dot.

So the well-known and established picture of spin-flip scattering as the driving force behind the Kondo-effect provides a clear understanding of the asymmetry-independence. However, the insensitivity of the ZBA to asymmetries observed here as well as in experiments[5, 6] is highly nontrivial, if viewed in the light of the Meir-Wingreen formula (2). Obviously, a simple translation of maxima in the equilibrium spectra $\rho_{\mathrm{Eq}}(\omega)$, to maxima in the *nonequilibrium* differential conductance $G(V)$ fails, and a full understanding of the bias dependence of many-body correlations is required. The voltage dependent renormalization of the transmission is essential for a correct description of the conductance as already pointed out[9]. In the Kondo regime, our non-perturbative many-body calculation out of equilibrium has revealed that the redistribution of spectral weight in $\rho(\omega, V)$[13] with bias occurs in such a way that the maximum in $G(V)$ is always found at $\Delta_K \approx \Delta_0$, irrespective of R and E_d.

Fig. 3(a) summarizes the results for Δ_K as function of the applied magnetic field for small interaction $U/\Gamma = 1$ as used in Fig. 1. The finite width of the ZBA determines the threshold field above which the splitting can be observed. Here, Kondo correlations are small, and the slope of Δ_K approaches $\frac{1}{2}(1 + \frac{1}{R})$ at large fields. The constant offset indicates the Hartree shift due to a finite U.

Fig. 3(b,c) displays the normalized peak position Δ_K/Δ_0 of the ZBA for a symmetric junction with $U/\Gamma = 4, 8$. In both cases, the zero-field extrapolation is consistent with the asymptotic value of the equilibrium Kondo regime[7], $\lim_{\Delta_0 \to 0} \Delta_K/\Delta_0 = \frac{2}{3}$. As in equilibrium[8], Δ_K/Δ_0 increases with field, and in the strongly correlated regime [panel (c)] this increase in rather slow due to the presence of typical Kondo-logarithms. In contrast to the equilibrium Kondo regime, however, a crossover to a non-universal behavior $\Delta_K/\Delta_0 > 1$ occurs at fields of the order of the charge scale, $\Delta_0 \sim \frac{U}{2} = 4\Gamma \approx 45T_K$. For $U = 4\Gamma$ [panel(b)], the charge fluctuations dominate the physics much earlier, and the crossover is observed already at $\Delta_0 \sim \Gamma \approx 3T_K$. We attribute this non-
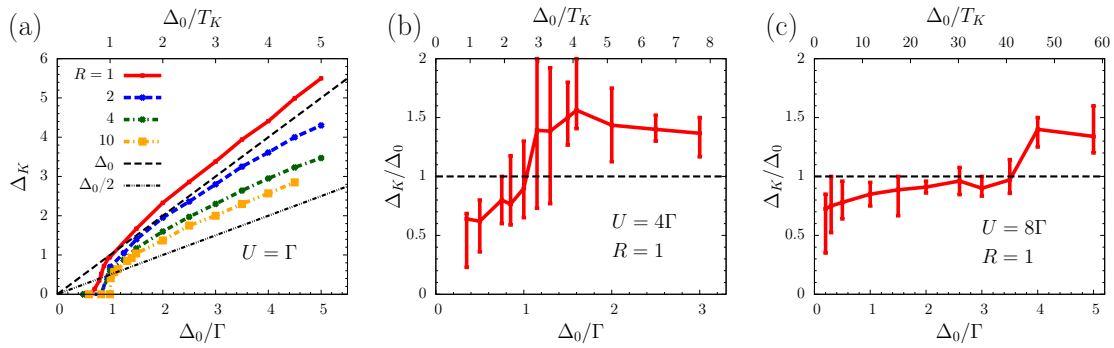
FIG. 3. (Color online) Position of the ZBA for particle-hole symmetric quantum dot as function of Zeeman energy for Coulomb interaction $U = \Gamma$ (a), 4Γ (b), and 8Γ (c). Panel (a) directly displays Δ_K calculated with the second Born approximation for various R, while panels (b) and (c) show the normalized position Δ_K/Δ_0 for a symmetric junction $R = 1$ obtained within the SNRG. The inaccuracies due to the numerical differentiation of $I(V)$ are indicated by the error bars in panels (b) and (c). The scale at the bottom measures Δ_0 in units of the coupling Γ, while the top scale is in units of the Kondo temperature T_K.

universal Zeeman splitting at large field to the nonequilibrium charge fluctuations.

We can distinguish two different regimes for strong correlations $U > \pi\Gamma$: (i) At fields and voltages below the charge excitation scale, Kondo correlations dominate leading to a renormalization of $\Delta_K < \Delta_0$, (ii) at high fields the Zeeman split Kondo peaks merge with the Hubbard side bands of $\rho(\omega, V)$ and the Kondo effect is destroyed. The physics is mainly driven by charge fluctuations accessible at large voltages and $\Delta_K > \Delta_0$ similar to the weakly correlated regime.

We have indeed reproduced the qualitative findings of experiments[6] and have identified the non-universal splitting as consequence of charge fluctuations. However, for $U/\Gamma \approx 8$ as estimated from experimental setups, the crossover in our calculations occurs at $\Delta_0/T_K \approx 40$ [panel (c)]. This is an order of magnitude larger than the values reported in experiments where the crossover typically occurs at $\Delta_0/T_K \approx 3 - 7$. We can reproduce such values only with a smaller Coulomb interaction $U/\Gamma = 4$. This might be hint for an additional transport mechanism present in experiments, which is not accounted for in the simple SIAM investigated here.

In summary, we have investigated the Zeeman splitting of the ZBA for weak, intermediate and strongly correlated nanodevices under true nonequilibrium conditions. In weakly correlated devices or generally at large bias the transport is driven by charge fluctuations, and the splitting strongly depends on the junction asymmetry. In contrast, for the Kondo-correlated ZBA present at large U/Γ our approach predicts an almost asymmetry-independent splitting, which is in accord with experiments and can be understood in a simple spin-flip scattering picture. Our theory explains the experimentally observed crossover[6] from $\Delta_K < \Delta_0$ to $\Delta_K > \Delta_0$ from spin to charge fluctuation driven transport, but also reveals shortcomings of an oversimplified description using only a single level due to the inaccurate scale for this crossover.

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